

Gasification of biomass wastes in an entrained flow gasifier: Effect of the particle size and the residence time

Juan J. Hernández^{a,*}, Guadalupe Aranda-Almansa^a, Antonio Bula^b

^a Universidad de Castilla-La Mancha, Departamento de Mecánica Aplicada e Ingeniería de Proyectos, Escuela Técnica Superior de Ingenieros Industriales (Edificio Politécnico), Avenida Camilo José Cela s/n, 13071 Ciudad Real, Spain

^b Universidad del Norte, Departamento de Ingeniería Mecánica, Km.5 Antigua Vía Puerto Colombia, Barranquilla, Colombia

ARTICLE INFO

Article history:

Received 22 July 2009

Received in revised form 7 January 2010

Accepted 24 January 2010

Keywords:

Biomass

Gasification

Entrained flow gasifier

Space residence time

Fuel particle size

ABSTRACT

Experimental tests in an entrained flow gasifier have been carried out in order to evaluate the effect of the biomass particle size and the space residence time on the gasifier performance and the producer gas quality. Three types of biomass fuels (grapevine pruning and sawdust wastes, and marc of grape) and a fossil fuel (a coal–coke blend) have been tested. The results obtained show that a reduction in the fuel particle size leads to a significant improvement in the gasification parameters. The thermochemical characterisation of the resulting char–ash residue shows a sharp increase in the fuel conversion for particles below 1 mm diameter, which could be adequate to be used in conventional entrained flow gasifiers. Significant differences in the thermochemical behaviour of the biomass fuels and the coal–coke blend have been found, especially in the evolution of the H_2/CO ratio with the space time, mainly due to the catalytic effect of the coal–coke ash. The reaction temperature and the space time have a significant effect on the H_2/CO ratio (the relative importance of each of these parameters depending on the temperature), this value being independent of the fuel particle size.

© 2010 Elsevier B.V. All rights reserved.

1. Introduction

Europe, as well as the whole world, must face up to a challenging energy scenario characterised by the following features [1–3]: a growing global energy demand (European energy demand is expected to grow 60% by 2030), rising of the energy dependency (to around 70% in the next 20–30 years) on oil and natural gas (frequently from politically instable producing regions), high prices and concerns on mid-term availability of fossil fuels, and the need of reducing the greenhouse gas emissions. In this sense, the European energy policies are focused in four main areas: the management of both internal demand and external supplies, a greater efficiency in the domestic market and the diversification of European energy supply sources [3].

Within the European energy strategy, the promotion and development of renewable energy play a major role [3]. In particular, the '20–20–20' targets establishing a 20% share of renewable energy in the EU energy consumption (along with a 20% reduction in greenhouse emissions and a 20% improvement in energy efficiency) by 2020 are remarkable [4]. Among renewable energies, the use of biomass as an energy resource entails environmental and socioeconomic benefits, such as waste disposal, zero net CO_2 emissions and social and economic development of rural areas [5,6]. Moreover, biomass is a geographically widespread and abundant resource [7]. Aware of the huge potential of

biomass in Europe, the European Commission adopted in 2005 a plan to increase the use of energy from forestry, agriculture and waste materials in heating, electricity and transport [3,8]. In consistency with the European policy, Spain developed a strategic plan on Renewable Energies, according to which 12% of the total energy consumption should come from renewable sources. The objectives for biomass were aimed at 1849 MW of electric energy in 2010 [9]. However, in 2007, only 396 MW of electricity (21.4% of the objective) were produced from biomass [10]. These data prove the urgent impulse that biomass technologies need in order to fulfil the objectives of the European and Spanish policies.

Among the different thermochemical processes currently available for biomass exploitation, biomass gasification (conversion of a carbonaceous feedstock into a gaseous energy carrier by partial oxidation at elevated temperature [11,12]) is one of the technologies that are receiving more attention from researchers and investors. Gasification appears as an attractive alternative to direct combustion, since it allows the reduction of storage and transport costs by means of the installation of small, low-cost and efficient gasifier-engine systems [13,14], as well as the recovery of available energy from low-value (biomass wastes and low-rank coals) materials, thereby reducing both the environmental impacts and the disposal costs [15]. The gas obtained, called producer gas or syngas, after cleaning and conditioning, can be used as a fuel in gas engines and turbines owing to its acceptable thermochemical combustion properties (flame speed and knock tendency) [13,16,17]. Gasification is also considered as a cleaner and more efficient technology than combustion, since it enables higher electric performances (30–32%

* Corresponding author. Tel.: +34 926 295300x3880; fax: +34 926 295361.

E-mail address: JuanJose.Hernandez@uclm.es (J.J. Hernández).

using gas engines compared to 22% achieved with a conventional Rankine cycle) [18], lower NO_x and SO_x emissions, and CO₂ capture [15]. However, biomass gasification must overcome some barriers before its commercial implementation. The main ones are the removal treatment of particles and tars, issues related to the production, logistics, and pretreatment of the biomass feedstock, and a better knowledge and understanding of the effect of the biomass properties and the gasifier operating conditions on the producer gas quality and the gasifier performance [19].

Within the currently available gasification technologies, entrained flow gasifiers constitute an interesting option owing to their commercial large scale availability and their high efficiency for the production of syngas [11,20]. These gasifiers operate at high temperatures (around 1200–1500 °C) and high heating rates, and require a finely reduced feedstock in order to achieve high levels of fuel conversion [20]. In addition, the high reaction temperature causes low tar formation but requires higher quality gasifier materials. According to Wei et al. [21,22], this type of gasifier offers the possibility of constant temperature in the reactor, higher heating rate and short but narrowly distributed residence times. However, current commercial entrained flow gasifiers are devoted mainly to coal and liquid fuels, that is, there exists little experience with biomass as feedstock [11]. An interesting alternative would be the use of biomass in existing, conventional coal-based entrained flow gasifiers. Actually, several options have been suggested in order to make biomass comply with the feeding demands of such systems: pulverisation (100 µm in size) or torrefaction. However, milling implies a great cost of pretreatment [20] and, hence, that makes biomass entrained flow gasifiers remain commercially unattractive [23]. As an alternative and due to the higher reactivity of biomass compared to that of coal, larger biomass particle size could be used leading to higher carbon conversion [11]. This would imply not only the reduction in the fuel pretreatment costs, but also the possibility of using available and inexpensive feeding systems such as screw feeders [20].

Fuel particle size, along with other fuel properties (moisture content, heating value, ultimate and proximate analysis) and gasifier operating conditions (gasifying agent, temperature, heating rate, biomass/air ratio, etc.), has been reported as one of the main parameters affecting the composition, quality and final applications of the producer gas [24,25]. Indeed, fuel particle size influences the time necessary for the gasification process to take place, as well as the adequate reactor size. It also plays an important role in all the successive reaction steps (fuel heating, reactant and product diffusion between the particle and the reaction atmosphere, and solid–gas reactions) which occur during the conversion of biomass into product gas.

A smaller particle size (related to a higher particle external surface area/volume ratio) enables a higher producer gas quality, a reduction in the reactor size or a lower space residence time to achieve a complete cracking of the heaviest and condensable fractions [18]. Wei et al. [22] studied the effect of the particle size in the pyrolysis process in a free fall reactor, and they concluded that smaller particles lead to an increase in the gas yield and a decrease in char and tar yields. Lv et al. [26], who performed an experimental study of air–steam gasification in a fluidized bed, concluded that a smaller particle size causes a higher carbon conversion and gas calorific value. Reed and Das [25], in an in-depth work on downdraft fixed bed gasifiers, stated that fuel particle size and shape determine the difficulty of fuel feeding, as well as its behaviour inside the reactor. Tinaut et al. [27] found that the maximum efficiency (represented by the biomass burning rate and the process propagation velocity) was obtained for smaller particle sizes and lower air velocities, owing to the higher fuel/air ratio in the gasifier. Chen et al. [28], who conducted a parametric study on pyrolysis/gasification in a fixed bed reactor, concluded that both a smaller fuel particle size and longer residence times resulted in higher gas yields. Similar results were obtained in a fluidised bed by Rapagnà and Latif [29], who found that the process is mainly controlled by the reaction kinetics for smaller particles, and, as the particle size increases, kinetic control gives way

to heat transfer control. Encinar et al. [30] suggested that fuel particle size affects the process velocity, and is related to mass and heat transfer. Chen and Gunkel [31], in a model of downdraft moving bed gasifiers, established that the larger the particle size, the lower its surface temperature, and thus, more heat is required for the reactions to take place. Mermoud et al. [32], who performed a numerical study of steam gasification in a charcoal particle, established that the minimum particle size for which diffusive effects are overcome (and thus, gasification rate remains constant) was between 0.2 and 1.8 mm.

As far as the space residence time (which is inversely related to the space velocity of the reactants) is concerned, this operational variable has influence upon the conversion and emissions of the process [33]. Wang and Kinoshita [34] performed a parametric study on biomass gasification from a kinetic model, and found that the conversion increased rapidly during the first 20 s of the process, and thereafter, chemical reactions started to proceed more slowly. Chen et al. [28] concluded that the space residence time of the volatile phase influenced positively on the pyrolysis gas yield. Xu et al. [35] showed that an increase in the space time leads to a rise in the efficiency of the gasification process at a dual fluidized bed reactor.

Most of the studies reported above have been focused on either fixed bed or fluidized bed reactors and have not considered the separate effect of the biomass particle size and the space residence time, both having a significant influence in the kinetics of the process and thus on the gasification efficiency. Thus, this paper, as a continuation of a previous work focused on the study of the effect of the biomass origin and the gasifier operating conditions on the gasification process [19], aims to shed light on the effect of the fuel particle size and the space residence time on the performance of an atmospheric entrained flow gasifier fuelled with different types of biomass (with interest in the southern regions of Europe), and a coal–coke blend. Given the relatively scarce literature reporting experimental work on biomass entrained flow gasifiers, the objectives are to achieve a better and a more comprehensive understanding of the gasification process, as well as to help establish the optimal operating conditions. The results obtained may also contribute to the development of entrained flow gasification as a feasible technology for biomass as feedstock.

2. Materials and methods

2.1. Gasification installation

The experimental tests have been carried out at the gasification equipment shown in Fig. 1. The pilot plant consists of a biomass feeding system (a lock hopper and a calibrated screw feeder) which enables to supply a controlled biomass flow. A crusher and a mill allow to grind the fuel prior to filling the lock hopper. The air used as gasifying agent comes from a compressor, and its pressure and volumetric flow have been measured and controlled in order to attain the required space residence time.

The entrained flow gasifier (operating at near atmospheric pressure) consists of an electric furnace with three independent temperature zones (7 kW each zone) which enable to keep the reaction temperature constant at the desired value. The furnace surrounds an alumina reaction tube (1.2 m length, inner diameter 60 mm, 7.5 mm thickness), which is the reaction chamber. Three R-type thermocouples have been used to measure and control the temperature of the three zones of the reactor. An ash–char hopper placed at the bottom of the tube allows to collect the sub-products of the gasification process (ash and char). The gasifier exit temperature (which gives an indication of how the process is taking place) is measured by means of a T-type thermocouple.

The producer gas generated in the process goes through a cooler and a fabric filter, which retains and allows to collect the rest of the particles flowing in the gas. A set of valves enables to lead the gas into a gas burner (or directly out through a chimney) or into a tar sampling

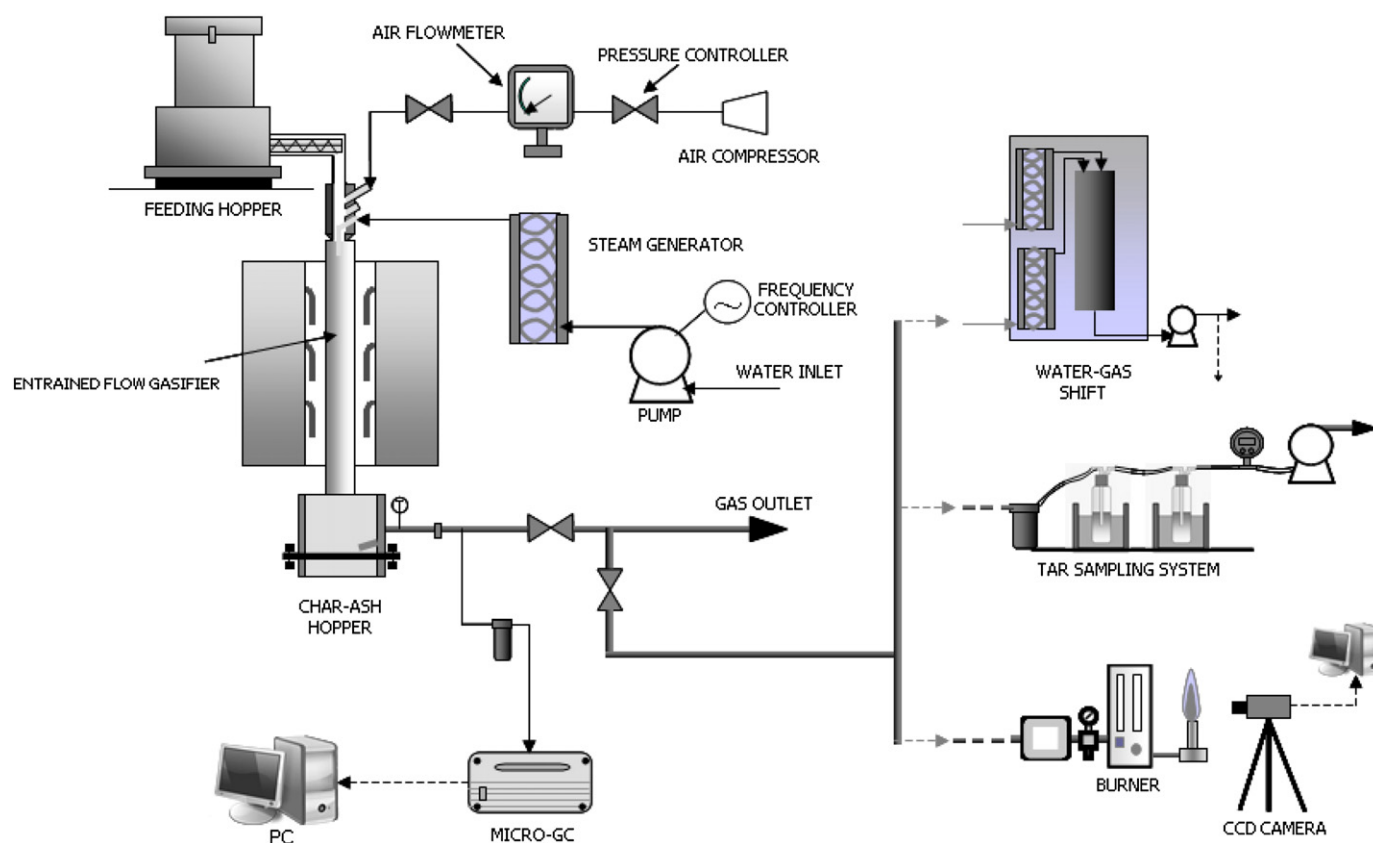


Fig. 1. Gasification pilot plant.

line which consists of a set of impingers immersed in a hot (40 °C) and a cold (−20 °C) bath, a gas mass flow-meter and a vacuum pump. However, and owing to the fact that the amount of tars can be considered negligible since the reactor temperature is very high, the tar sampling line has not been used in the present work.

A sampling line including a small particle filter and a pump allows to measure in-line (every 2 min) the producer gas composition by means of a micro-GC (Agilent 3000), equipped with a thermal conductivity detector (TCD) and two columns (a molecular sieve column to detect CO, H₂, CH₄, N₂ and O₂, and a Plot-U column to measure CO₂ and C₂H₆).

2.2. Thermochemical characterisation

The biomass fuels tested have been chosen for being abundant and representative of agricultural (grapevine pruning), forestry (pine sawdust) and industrial (dealcoholised marc of grape) wastes in the inland regions of Spain. The description of the wastes tested, as well as their origin and potential resources, has been reported in a previous work [19]. On the other hand, the coal-coke blend tested (coming from a low-rank autochthonous coal and a residue from oil refining

process) is the fuel used at the ELCOGAS GICC power plant, located in Puertollano (Spain) [36,37].

Prior to the gasification experimental tests, thermochemical analysis of the biomass fuels was carried out in order to determine the effect of the fuel composition on the producer gas quality. The proximate analysis of the fuels was obtained by means of a TA Instruments Q500 Thermo-Gravimetric Analyser (TGA) (the experimental procedure having been described in references [14,19]). On the other hand, the ultimate analysis was performed with a Leco CHNS-932 (according to the CEN/TS-15104 and CEN-TS-15289 procedures [38,39]). Fuel higher heating value (HHV) was obtained with a Parr calorimetric vessel (according to the UNE-164001-EX [40]). Lower heating value (LHV) was subsequently calculated from HHV and the biomass hydrogen content. Simultaneously to all analyses, the moisture content of the samples was determined with an A&D MX-50 moisture analyzer in order to transform the obtained thermochemical properties to a dry basis. The same experimental methodology was used for the characterisation of the gasification char-ash residues. The thermochemical properties of the fuels tested are shown in Tables 1 and 2.

As shown in Table 1, significant differences between the fossil coal-coke blend and the biomass fuels can be observed. Firstly, biomass has a

Table 1
LHV, ultimate and proximate analysis (dry basis) of the fuels tested.

Fuel	Moisture (wt.%)	LHV (MJ/kg)	Ultimate analysis (wt.%)					Proximate analysis (wt.%)		
			C	H	N	S	O ^a	Volatile matter	Fixed carbon ^b	Ash
Grapevine pruning	6.28	17.57	48.10	5.44	0.79	0.04	41.32	77.45	18.25	4.32
Sawdust wastes	5.75	18.62	49.99	5.81	0.10	0.18	43.52	85.26	14.34	0.40
Dealcoholised marc of grape	7.79	18.93	51.20	5.53	2.48	0.17	33.62	65.70	27.28	7.00
Coal-coke (50 wt.% each)	1.90	22.50	61.51	3.13	1.50	2.94	5.23	14.81	59.49	25.70

^a By difference (ash-free).

^b By difference.

Table 2
Stoichiometric biomass/air ratios (kg dry fuel/kg air) for the fuels tested.

Fuel	Empirical formula	F_{stoic}
Grapevine pruning	$\text{CH}_{1.36}\text{O}_{0.64}\text{N}_{0.014}\text{S}_{0.003}$	0.198
Sawdust wastes	$\text{CH}_{1.39}\text{O}_{0.65}\text{N}_{0.017}\text{S}_{0.0013}$	0.197
Dealcoholised marc of grape	$\text{CH}_{1.29}\text{O}_{0.49}\text{N}_{0.041}\text{S}_{0.0012}$	0.175
Coal-coke (50 wt.%)	$\text{CH}_{0.61}\text{O}_{0.064}\text{N}_{0.021}\text{S}_{0.0018}$	0.114

much higher volatile content than coal-coke. As stated by some authors [20], a high volatile content is directly related to the fuel reactivity (how fast the fuel is converted into gas) and, hence, results in higher fuel conversion. Therefore, the obtained data show that biomass is much more reactive than carbon-coke, which is in agreement with other authors [41,42]. In addition, the ash content of coal-coke, as well as the sulphur content, is higher than that of the biomass. Although the marc of grape is the biomass fuel with the lowest volatile content, its higher ash amount when compared to the rest of biomasses (mainly the significant potassium content (see Table 6)) could improve the expected gasification behaviour. Actually, several authors [11,19,22,33,41,43–45] point out that the inorganic elements present in the biomass ash (namely K, Na, Fe, and Ca) could act as catalysts for the pyrolysis, combustion, and gasification processes. Likewise, porosity and pore distribution must be also considered when determining the reactivity of a fuel [41].

Ultimate analyses show that carbon, hydrogen, sulphur and nitrogen content of biomass fuels are similar, although the relatively higher nitrogen content of the marc of grape compared to the rest of biomass samples is remarkable. However, sulphur contents are very low for all the biomass fuels tested. Table 2 presents the empirical formula and the stoichiometric fuel/air ratio (F_{stoic}) for the different fuels used. These data are derived from those obtained in the ultimate analysis. As can be observed, all the biomass fuels have similar stoichiometric fuel/air ratios, and these values are higher than that for the coal-coke blend (that is, biomass needs less oxygen to get completely oxidised). The F_{stoic} value has been used to specify the relative fuel/air ratio (F_{rg}) of the gasification tests, which is defined with respect to the stoichiometric one.

2.3. Experimental schedule

As can be observed in sections below, several experimental sets have been performed. In the first one (Table 3 and Section 3.1), the dealcoholised marc of grape has been used as gasification fuel in order to study the effect of the particle size. This fuel has been chosen because of its abundance, good thermochemical properties and its relatively easy grindability. The fuel was grinded and sieved at different particle diameters, and homogenised prior to the gasification tests. The experimental schedule was designed so that the rest of operating conditions (reaction temperature and relative fuel/air ratio) could be kept as constant as possible. As a complementary study, thermochemical characterisation (ultimate and proximate analyses and heating value) of the resulting char-ash was carried out in order to achieve a better understanding of the reactions taking place during the conversion process.

Table 3
Experimental conditions for the study of the effect of the fuel particle size.

Experimental conditions							
Fuel	d_p (mm)	T (°C)	p (bar)	F_{rg}	\dot{m}_f (kg/h)	\dot{m}_a (kg/h)	FC (%)
Dealcoholised marc of grape	8	1050	3	4.25	1.67	2.29	57.54
	4			3.91	1.51	2.26	64.43
	2			3.88	1.58	2.38	73.83
	1			3.98	1.44	2.11	80.35
	0.5			4.25	1.49	2.04	91.36

Table 4
Experimental conditions for the study of the effect of the space residence time.

Experimental conditions						
Fuel	T (°C)	d_p (mm)	F_{rg}	\dot{m}_f (kg/h)	\dot{m}_a (kg/h)	t_r (s)
Grapevine pruning	1050	<0.5	3.20	1.09	1.70	1.92
			2.92	1.22	2.08	1.57
			3.11	1.40	2.24	1.46
Sawdust wastes	1050	<0.5	3.15	1.52	2.40	1.36
			1.88	0.58	1.70	1.92
			1.99	0.75	2.08	1.57
			1.75	0.71	2.24	1.46
Dealcoholised marc of grape	1050	<0.5	2.05	0.89	2.40	1.36
			4.01	1.17	1.70	1.92
			3.79	1.35	2.08	1.57
			3.93	1.51	2.24	1.46
Coal-coke (50 wt.%)	1050	<0.5	3.79	1.56	2.40	1.36
			8.04	1.23	1.70	1.92
			7.32	1.37	2.08	1.57
			7.92	1.71	2.40	1.36

For the second experimental set (Table 4 and Section 3.2), the study was focused on the effect of the space residence time for several fuels (three types of biomass and a residual coal-coke blend). As in the previous schedule, the relative fuel/air ratio was kept as constant as possible. The last experimental schedule (Table 5 and Section 3.3) has been designed with the aim of studying the combined effect of reaction temperature and space residence time, both parameters influencing the reaction rate of the gasification process.

Prior to the experimental tests, all fuels were milled, ground and sieved to the required particle size (below 0.5 mm when studying the effect of the space time). Once the fuel pretreatment was carried out, a representative sample of each fuel was taken in order to conduct thermochemical characterisation tests.

Before each run, the lock hopper was filled with a weighed amount of biomass. The furnace was set on the selected temperature (1050 °C in all cases, except for the tests to study the effect of both the temperature and the space time). Once the reactor tube reached the selected temperature, air (the gasifying agent used for all the tests) was introduced according to the selected flow and pressure. The reference air flow was set as 2 Nm³/h at 3 bar. According to such reference, the space residence time was changed (Sections 3.2 and 3.3) by modifying the pressure (and hence, the flow rate) of the gasifying agent. Then, biomass was fed into the reactor at a volumetric flow set by means of a calibrated screw feeder, but keeping the relative fuel/air ratio constant (thus allowing to compare the results obtained for different space residence times). In that moment, the gasification run started. Producer gas samples were taken and analysed every 2 min with a gas micro-chromatograph (Agilent 3000). The micro-GC operating conditions are shown in Table 6.

The temperature at the exit of the gasifier and the air flow were periodically registered. After 15–20 min, when the gas composition remained constant and steady state was achieved, the run was finished.

Table 5
Experimental conditions for the study of the combined effect of the reaction temperature and the space residence time.

Reference	Experimental conditions					
	Fuel	T (°C)	\dot{m}_f (kg/h)	\dot{m}_a (kg/h)	F_{rg}	t_r (s)
Lapueta et al. [19]	Grapevine pruning	750	1.27	2.31	3.1	1.82
		850	1.27	2.39	3.0	1.61
		1000	1.27	2.34	3.1	1.45
		1150	1.27	2.30	3.2	1.32
This study		750	1.56	2.08	3.5	2.03
		850	1.56	2.08	3.5	1.85
		950	1.56	2.08	3.5	1.70
		1050	1.69	2.08	3.8	1.57

Table 6
Micro-GC operating conditions.

	Column 1	Column 2
Carrier gas	He	Ar
Injection temperature (°C)	100	70
Column temperature (°C)	110	70
Column pressure (psi)	30	25
Run time (s)	120	120

At this moment, the final content of the lock hopper was weighed in order to calculate the biomass mass flow rate. The furnace was set to ambient temperature. When the temperature was low enough to ensure a safe operation, char and ash contained in the bottom hopper and the fabric filter were collected, weighed, and properly sampled and stored for subsequent characterisation analyses.

The main parameters shown in this work describing the gasification process are the following (all the parameters refer to the biomass flow rate on a dry ash-free (d.a.f.) basis):

- Producer gas lower heating value, LHV_{pg} (MJ/kg): it has been calculated from the producer gas composition (on a dry basis) and the corresponding value for the combustible species (CO, H₂ and CH₄).
- Gas yield, GY (kg dry gas/kg biomass d.a.f.): dry producer gas flow rate with respect to the biomass flow rate.
- Cold gas efficiency, η_g (%): calculated as the ratio between the producer gas energy content (based on its LHV) and the biomass energy content (on dry ash-free basis) at standard conditions (273 K, 1 atm).
- Hydrogen/carbon monoxide ratio in the producer gas, H₂/CO.

3. Results and discussion

3.1. Effect of the fuel particle size

Table 3 shows the experimental conditions for the tests performed to analyse the effect of the fuel particle size, \dot{m}_f and \dot{m}_a being the biomass flow rate and the air flow rate, respectively. All tests were carried out at 1050 °C and at a relative F_{rg} value around 4, which is typical in gasification processes. Fuel conversion data (defined as the fuel proportion converted into producer gas) were obtained from the weighing of the char–ash residue collected after each test, as shown in Eq. (1) (where m_{char} and m_f are the char–ash residue produced and the fuel mass used in each test respectively).

$$FC(\%) = \left(1 - \frac{m_{char}}{m_f}\right) \cdot 100 \quad (1)$$

Fig. 2 shows the results obtained. It can be seen that the concentration of all the combustible species (CO, H₂ and CH₄) increases as the fuel particle size reduces, whereas CO₂ concentration slightly diminishes. As far as fuel conversion is concerned, it increases (57.5% for 8 mm diameter particles) when reducing the fuel particle size, reaching a value as high as 91.4% for 0.5 mm diameter.

Fig. 3 plots the results obtained for gas yield, LHV and η_g . As can be seen, the combined effect of a higher heating value and a nearly constant gas yield as the fuel particle size decreases leads to higher cold gas efficiency values. These results are consistent with those obtained in literature [11,22,26,29,30,33,34]. The smaller the fuel particle size, the more effective are mass and heat transfer since the

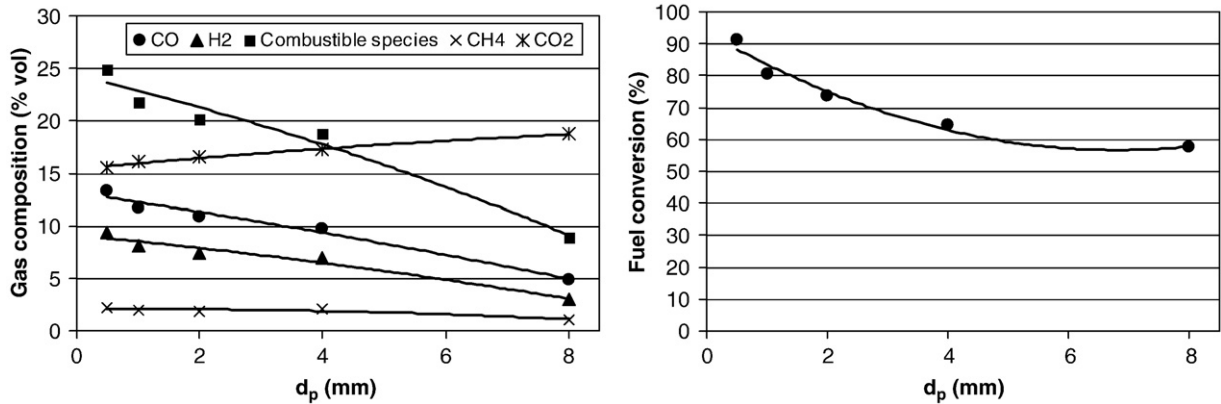


Fig. 2. Effect of the fuel particle size on the producer gas composition (left) and the fuel conversion (right).

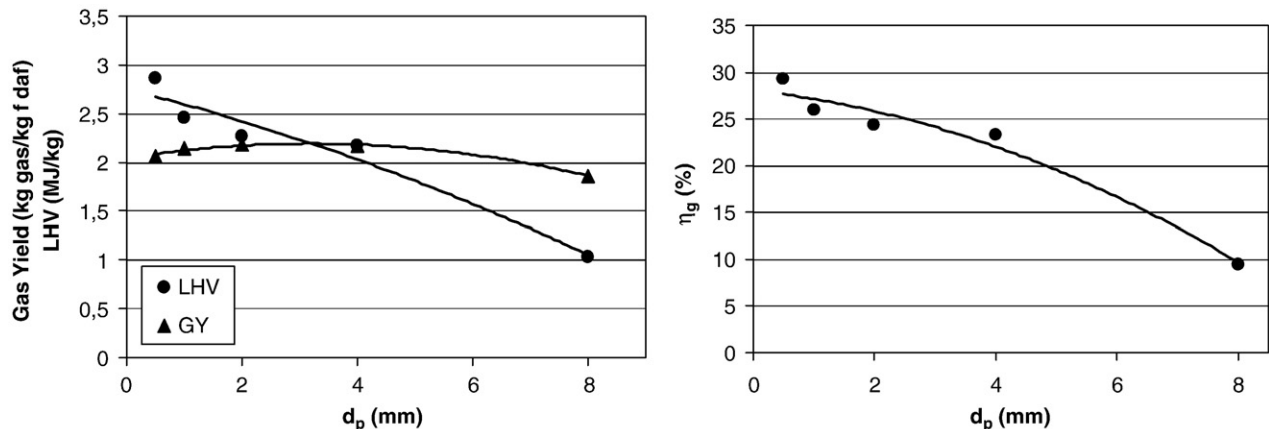


Fig. 3. Effect of the fuel particle size on LHV, GY (left) and η_g (right).

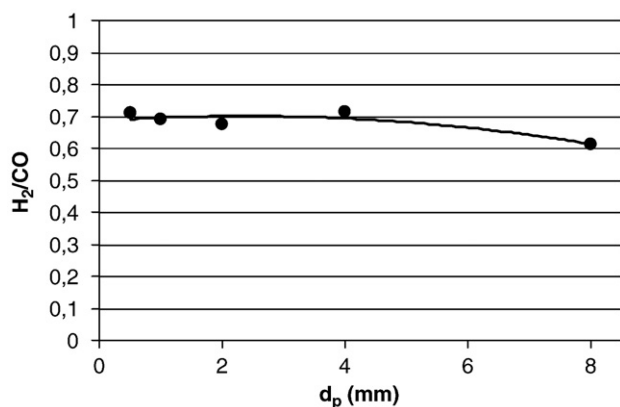


Fig. 4. Effect of the fuel particle size on the H₂/CO ratio.

particle external surface area/volume is higher and the char formed during pyrolysis is expected to be more porous owing to a higher volatile release (in agreement with the results obtained by Babu and Chaurasia, which concluded that a lower time is required for the completion of pyrolysis when the particle size decreases [46]). Therefore, the reactivity of the remaining char increases, and thus the gasification reactions take place to a higher extent. On the other hand, mass and heat transfer are improved (lower diffusion resistance coefficients) when diminishing the particle size, and chemical kinetics could become the rate-controlling factor. When reaction controls the process, reaction rate grows exponentially with the temperature and with the increase of the external surface area/volume ratio. The uniform temperature reached in the particle allows the reaction to take place throughout the particle, not only in its surface area (since the internal heat transfer conduction resistance, and thus, the temperature gradient inside the particle is reduced), and thus leading to an upgrading of the producer gas quality [22]. Fig. 4 shows that the H₂/CO ratio remains almost constant when changing the particle size (around a value of 0.7).

The gasification tests presented above were complemented with a thermochemical characterisation study of the char–ash residue obtained. The aim is to achieve a better comprehension of the reaction stages taking place during the whole conversion process depending on the fuel particle size. Figs. 5–7 show the results obtained in the proximate and ultimate analyses and the lower heating value. As can be observed, as the fuel particle size is reduced, the release of volatile

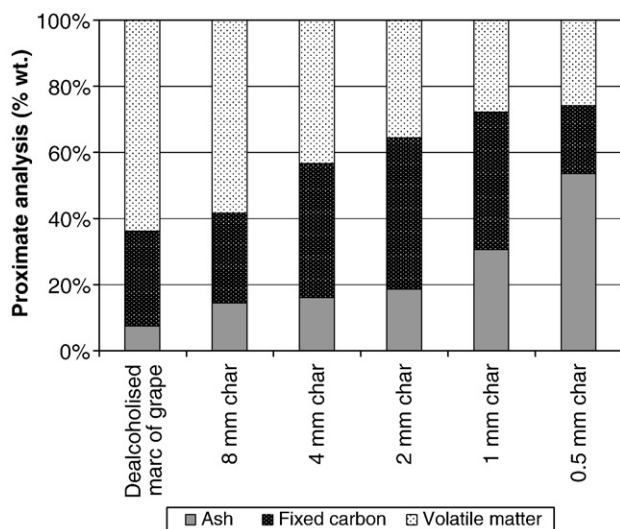


Fig. 5. Proximate analysis of char–ash residue obtained from different fuel particle sizes (dry basis).

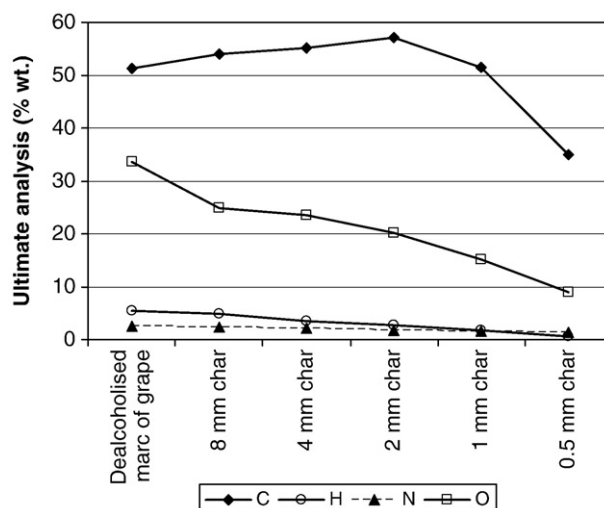


Fig. 6. Ultimate analysis of char–ash residue obtained from different fuel particle sizes (dry basis).

matter during the pyrolysis stage and the particle carbonisation gradually increase (Fig. 5), leading to a lower volatile content in the residue. Similarly, the ash content also rises, although at a slower rate. This indicates that pyrolysis reactions are enhanced as the particle size decreases. Nevertheless, for fuel particles below 1 mm, char gasification reactions start to take place to a greater extent, as can be observed by a sharp increase in the ash content (related to a higher particle conversion) and a proportional reduction in the fixed carbon content in the residue.

Fig. 6 displays the evolution of the elemental composition of the residue as the fuel particle diminishes, as well as the comparison with the composition of the original fuel. As can be seen, carbon content slightly increases as the fuel particle size decreases (which is associated with the increase in the char heating value, as shown in Fig. 7), suggesting a slow and progressive carbonisation of the particle, whereas hydrogen and nitrogen contents are reduced (indicating that the volatile release is favoured). However, between 1 and 0.5 mm, the carbon content (and thus the char heating value) suffers a sharp reduction, which indicates that for fuel particles below 1 mm diameter not only are pyrolysis reactions enhanced, but also are the char gasification ones, hence improving fuel conversion levels (as shown in Fig. 2).

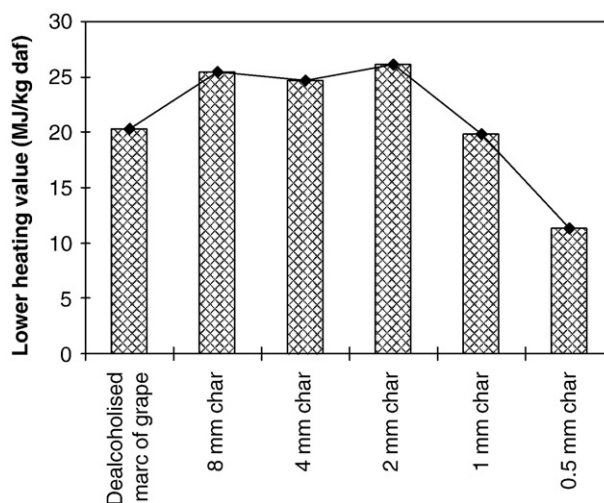


Fig. 7. Lower heating value of char–ash residue obtained from different fuel particle sizes (dry, ash-free basis).

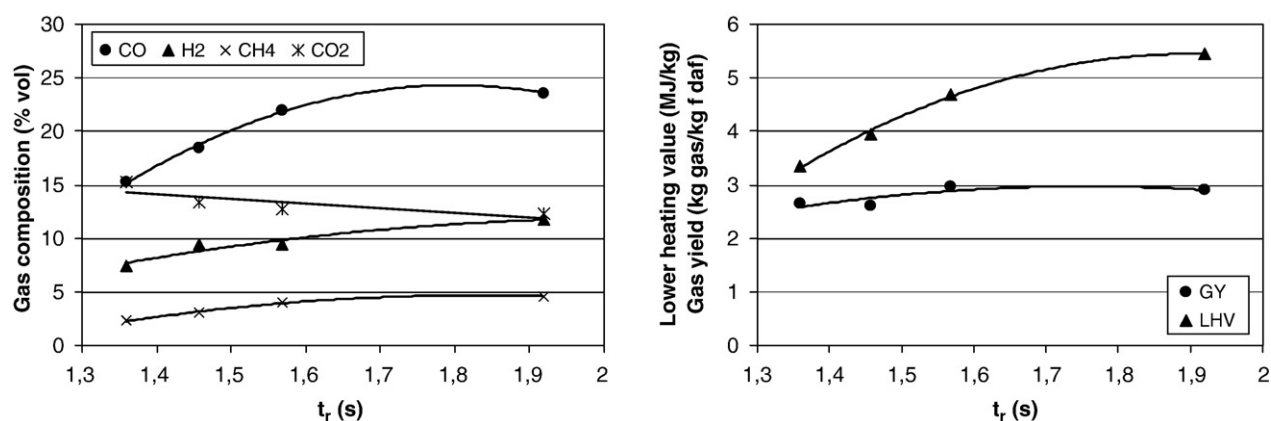


Fig. 8. Effect of t_r on the gas composition (left), LHV and GY (right) for grapevine pruning wastes.

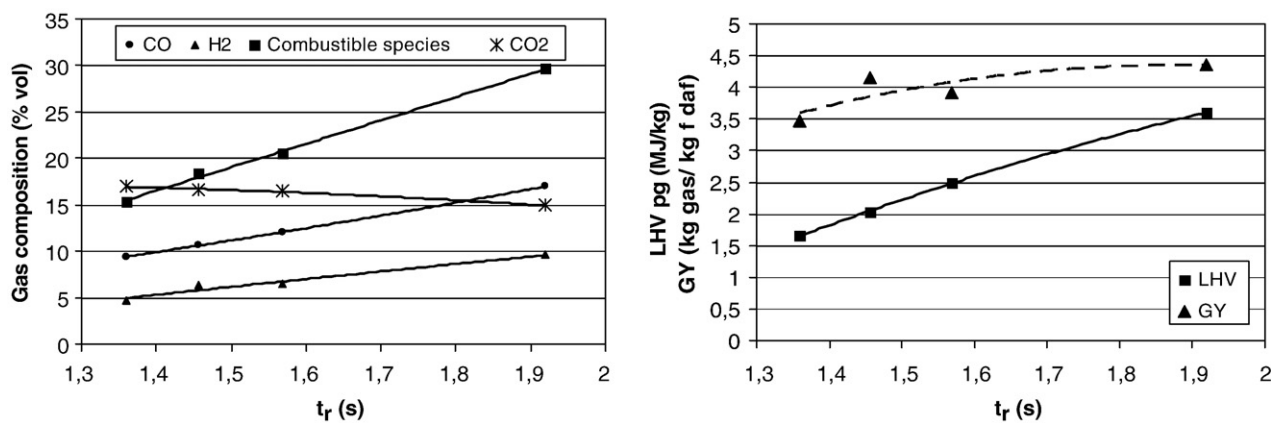


Fig. 9. Effect of t_r on the gas composition (left), LHV and GY (right) for sawdust wastes.

3.2. Effect of the space residence time

Table 4 presents the experimental tests performed to analyse the effect of the space residence time (t_r , defined as the reactor volume divided by the air volumetric flow rate) for different types of biomass while keeping the reaction temperature (1050 °C) and the relative fuel/air ratio (F_{rg}) constant. Due to the different fuel densities and the limitations in the volumetric screw feeder rate, the F_{rg} values achieved depend on the type of biomass. That is the reason why, unfortunately, the effect of the space residence time can only be seen separately for each fuel. Figs. 8–13 show the results obtained for the four fuels tested. From a qualitative point of

view, the effect of the space residence time (which is limited to very short values in an entrained flow gasifier) shows similar trends for all the biomass fuels.

As can be seen in Figs. 8–11, an increase in t_r causes an improvement in the producer gas quality since all the combustible species (CO, H₂, and CH₄) increase their concentration in the producer gas. These results are consistent with those obtained by Wang and Kinoshita [34], and can be explained by the closer approach to equilibrium values as the space time increases. As for CO₂, it slightly decreases due just to the increase of the combustible species, since in all the cases the air flow (and thus the oxygen available for the reactions) was kept nearly constant. As a

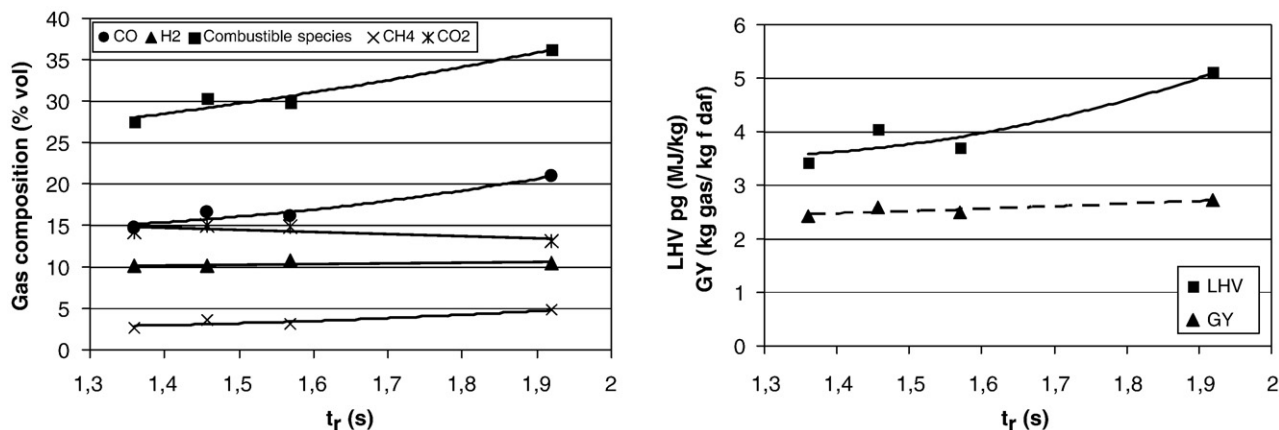


Fig. 10. Effect of t_r on the gas composition (left), LHV and GY (right) for dealcoholised marc of grape.

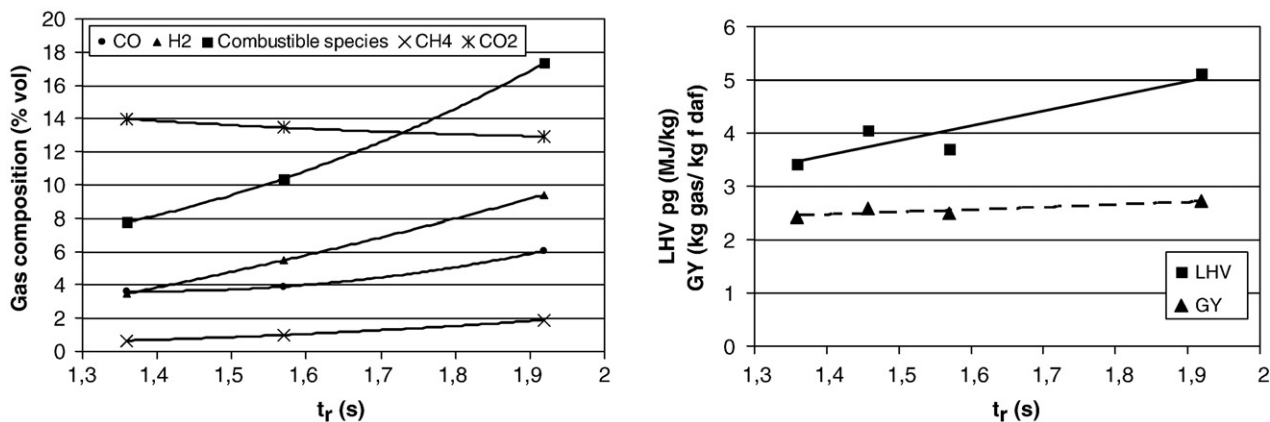


Fig. 11. Effect of t_r on the gas composition (left), LHV and GY (right) for the coal-coke blend.

consequence of this, the gas heating value increases, this effect being less significant in the case of the dealcoholised marc of grape. Both maximum CO and H₂ values were obtained for grapevine pruning wastes (23.6% vol. CO and 11.8% vol. H₂ at 1.9 s). CH₄, in general, remained fairly constant in all the cases, except for the sawdust wastes where it slightly rose up.

On the other hand, the gas yield (GY) rises slightly in the case of grapevine pruning wastes and dealcoholised marc of grape, although this increase is more significant in the case of sawdust and coal-coke blend. As far as the performance of the fuels tested is concerned, it has been proved that, even using higher F_{rg} values, the quality of the gas obtained from coal-coke is much lower than that obtained from biomass (17% of combustible species from the former compared to 30–40%

obtained from the latter). These figures can confirm the statement about the higher reactivity of biomass compared to that of coal (as mentioned in Section 2.2).

The similar effect of the space residence time on LHV and GY (the former increasing while the latter remains almost constant) makes cold gas efficiency increase in all cases (Fig. 12), in agreement with the results obtained by Xu et al. [35]. Grapevine pruning and sawdust exhibit the sharpest increase in η_g with the space time. The cold gas efficiency reaches 40% for the coal-coke blend compared to values as high as 70–89% for biomass at a space time of 1.9 s.

For the reaction temperature considered in this section (1050 °C), the space time has little effect on the H₂/CO ratio for the biomass fuels (Fig. 13), showing a very slight decrease just for the marc of grape.

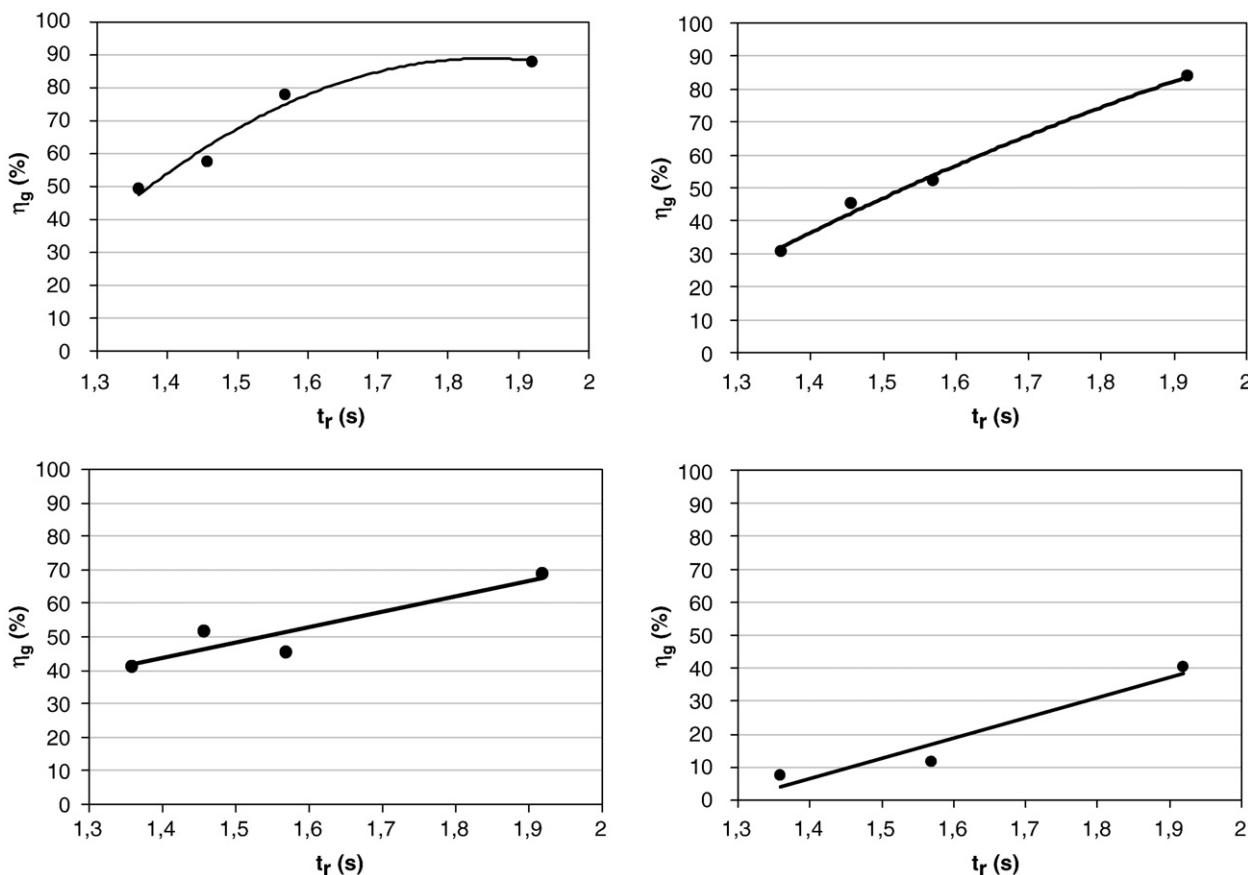


Fig. 12. Effect of t_r on η_g for grapevine pruning wastes (upper left), sawdust wastes (upper right), dealcoholised marc of grape (down left), and coal-coke (down right).

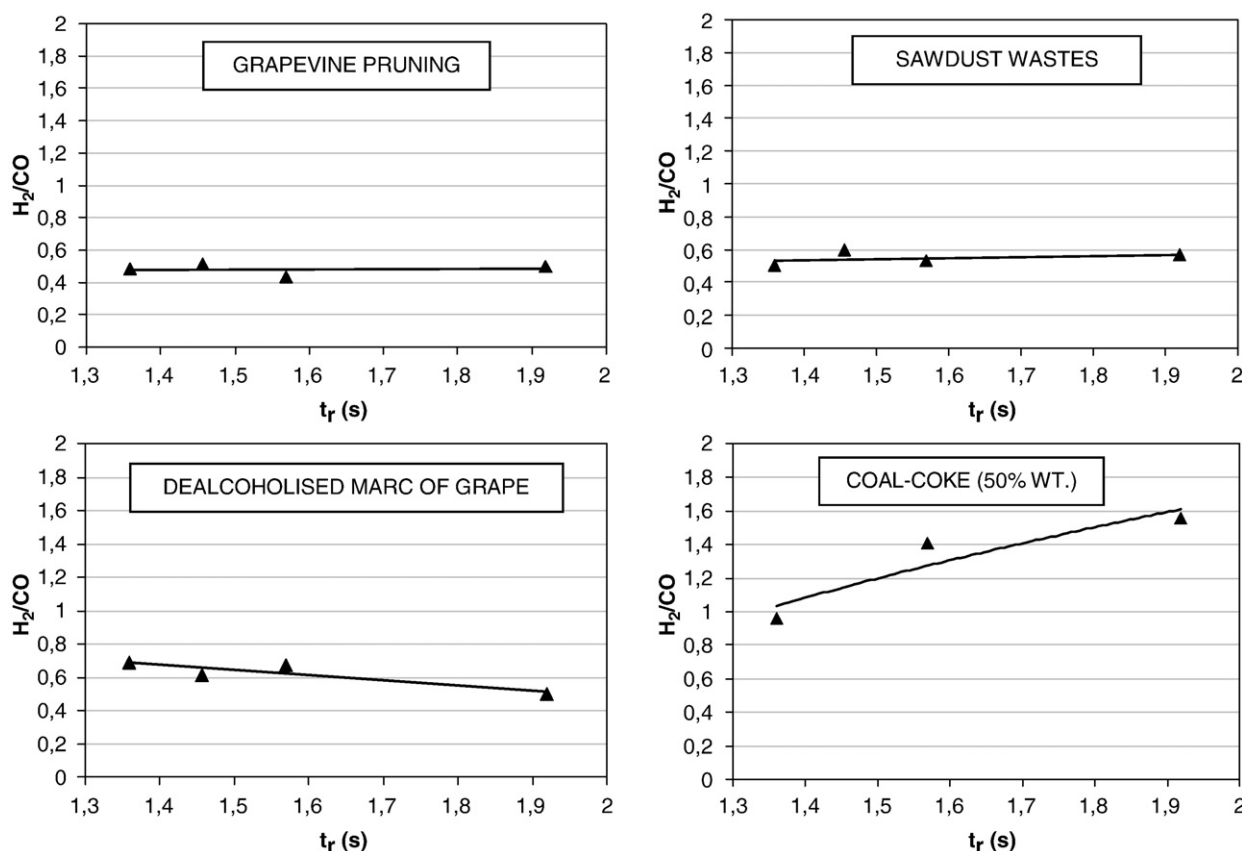


Fig. 13. Effect of t_r on the H_2/CO ratio for grapevine pruning (upper left), sawdust (upper right), dealcoholised marc of grape (down left) and coal-coke (down right).

However, the different behaviour of this ratio is relevant in the case of coal-coke, not only because of its higher values (between 0.85 and 1.56 for coal compared to those around 0.5 for biomass) but also because of their opposite trend, since an increase of the space residence time causes a rise in H_2/CO ratio. The key to this different behaviour might be the different molecular structure between biomass and coal, as well as their different ash composition. In this sense, Ye et al. [47] suggested that coal gasification rates strongly depend on the inorganic matter content, and that reactivity of a low-rank coal was related to the amount of inorganic constituents. Sutton et al. [48] state that the difference of reactivity between coals is a function of the ash content. Table 7 reports the composition of ash obtained from the marc of grape and that of coal-coke (in both cases, ash samples were obtained according to the UNE-CEN/TS 14775 EX procedure [49], and the chemical analysis was performed by using a X-ray fluorescence spectrometer). As can be seen, not only has the coal-coke blend a significantly higher amount of ash than biomass (see Table 1), but also this ash is composed of a higher quantity of Al, Si, Fe, and Ni, whereas lower content of K, and Ca. Iron, along with chromium, is a well-known commercial catalyst for the high-temperature water-gas shift process [50]. On the other hand, Al and

Zn (along with Cu) are commercially used as catalysts for the low-temperature water-gas shift process [51]. Likewise, Zn is reported to increase the hydrogen fraction when used as catalyst [48], being 8 times more effective in the H_2 production than other additives. Ni (18 times greater in ash from coal-coke compared to biomass) is a widely used catalyst for hydrocarbon, methane, and tar steam reforming [48], also used for the adjustment of syngas composition. On the other hand, higher t_r values (related to lower gas space velocities) favour gasification processes (such as tar reforming, and water-gas shift reaction) [52,53]. Therefore, the catalytic action of Fe, Ni, Al and Zn present in ash from coal-coke (which probably enhances water-gas shift reaction), along with longer space times inside the reactor, might be responsible for the higher and increasing H_2/CO trends found in this work.

3.3. Combined effect of the space residence time and the reaction temperature

As a complementary study, the combined effect of the reaction temperature and the space time has been studied. The reaction temperature influences the heating rate, the rate and equilibrium constants,

Table 7
Ash chemical analysis (wt.%) of dealcoholised marc of grape and coal-coke.

Fuel	Concentration (%)											
	Na ₂ O	MgO	Al ₂ O ₃	SiO ₂	P ₂ O ₅	SO ₃	K ₂ O	CaO	TiO ₂	MnO	Fe ₂ O ₃	NiO
Dealcoholised marc of grape	0.53	3.77	2.08	7.53	6.95	4.97	29.10	22.53	0.14	0.06	1.40	0.005
Coal-coke	0.46	0.78	25.1	52.05	0.07	2.04	2.34	4.41	0.64	0.04	4.40	0.09
	Ga ₂ O ₃	GeO ₂	As ₂ O ₃	Rb ₂ O	SrO	Y ₂ O ₃	ZrO ₂	BaO	CeO ₂	PbO	CuO	ZnO
Dealcoholised marc of grape	0	–	–	0.02	0.16	0.001	–	0.04	–	–	0.08	0.02
Coal-coke	0.005	0.005	0.01	0.01	0.01	0.006	0.02	0.04	0.06	0.04	0.01	0.08

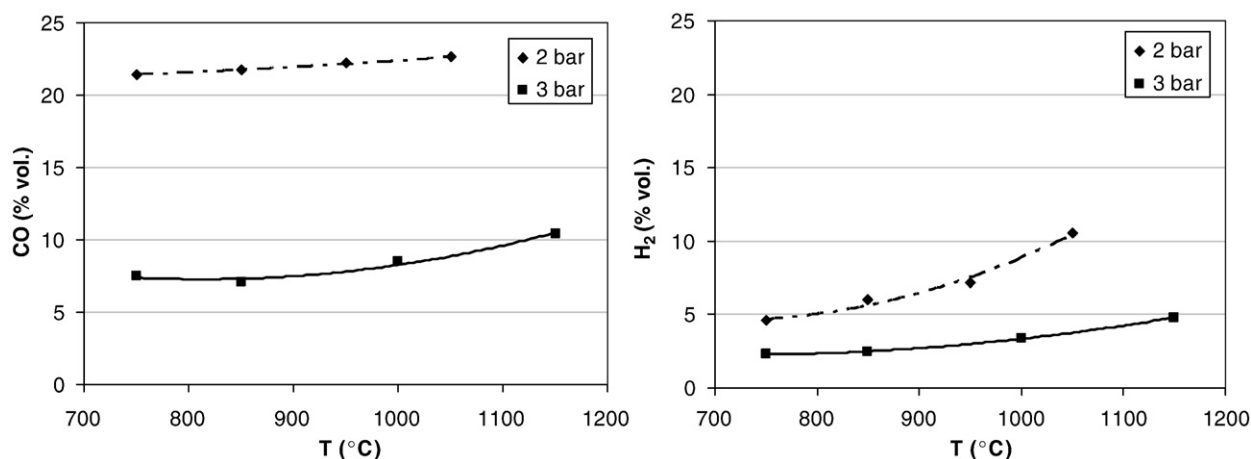


Fig. 14. Effect of the reaction temperature and the residence time on the producer gas CO (left) and H₂ (right) content for grapevine pruning wastes.

and the space residence time (due to the changes in the producer gas density), which in turn determine the products distribution [22,34].

Table 5 presents the experimental schedule for this study. Two data sets have been considered: one of them, performed for this study at a lower air pressure (2 bar), and thus a lower air spatial velocity and higher space time, and data taken from a previous work [19], in which the effect of the reaction temperature was studied at a higher air pressure (3 bar). In all cases, the changes in t_r are due exclusively to the change in the air density inside the reactor. Anyway, for lower air velocities, the space time is kept higher for all temperatures studied, as can be seen in the table. The slight difference between F_{rg} values in both data sets can be considered negligible.

The obtained results are depicted in Figs. 14 and 15, where it can be seen that the combined effect of an increase in both reaction temperature and space time leads to an increase in the CO and H₂ content (and thus, the producer gas LHV), and in the cold gas efficiency, in agreement with trends obtained by Wei et al. [21]. It can also be observed that, for the same temperature, longer space residence time affects the H₂ production to a greater extent than that of CO (Fig. 14). However, the effect of t_r on the H₂/CO ratio depends on the temperature of the process, this ratio decreasing when t_r increases for a temperature lower than 1000 °C while it rises at higher temperatures. This result is in agreement with that shown in Fig. 15, in which the H₂/CO ratio from grapevine pruning gasification remains constant with t_r at a temperature close to 1050 °C. The temperature at which this inflection point happens is likely to vary depending on the fuel, although this is planned to be studied in future works.

4. Conclusions

Several experimental schedules in an atmospheric entrained flow gasifier have been carried out in order to determine the effect of the fuel particle size (d_p) and the space residence time (t_r) on several gasification parameters, such as the producer gas composition (in particular, the CO and H₂ content), the gas heating value, the gas yield and the cold gas efficiency. Different types of biomass (agricultural, forestry and industrial wastes) with a high interest in the southern regions of Europe have been tested and the results have been compared to those obtained for a conventional fossil fuel (a coal-coke blend). The main conclusions obtained are the following:

- A reduction in the fuel particle size leads to an improvement in the gas quality (represented by an increase in the combustible species), and thus to a higher producer gas heating value. Cold gas efficiency, H₂/CO ratio and fuel conversion are also enhanced. Maximum fuel conversion was obtained for the smallest particle size tested (0.5 mm).
- Thermochemical characterisation of the char-ash residue shows that as the fuel particle size is reduced, the release of volatile matter during pyrolysis stage, along with particle carbonisation, gradually increase, suggesting that pyrolysis reactions take place to a greater extent. However, for fuel particles below 1 mm, char gasification reactions start to become more relevant, contributing to the improvement of the fuel conversion and the producer gas composition.
- Longer space residence time inside the reactor (achieved by means of lower air velocities) causes significant benefits for the gasification

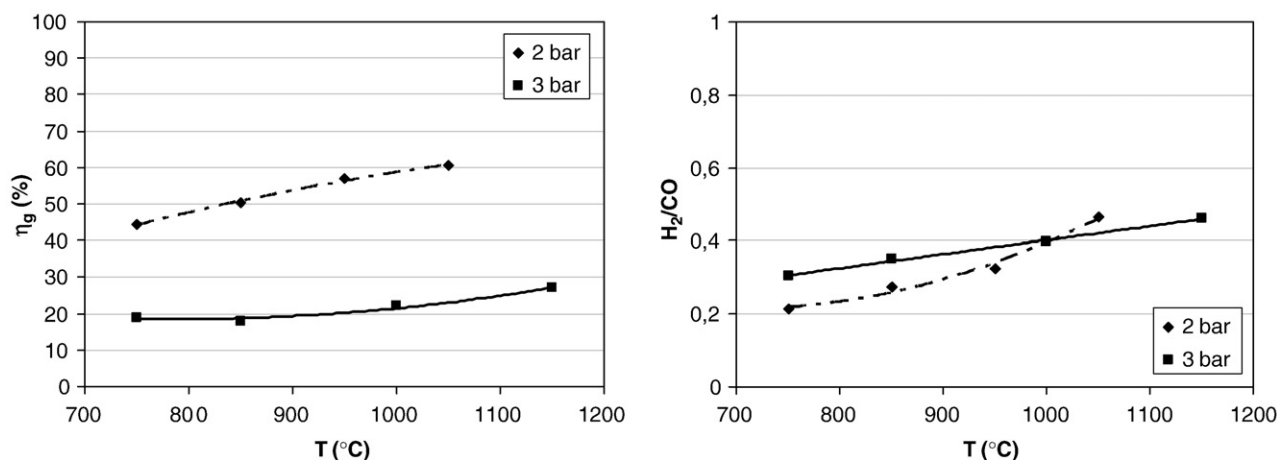


Fig. 15. Effect of the reaction temperature and the residence time on the η_g and the H₂/CO ratio for grapevine pruning wastes.

process, since all the parameters (CO and H₂ content, gas lower heating value, cold gas efficiency, and fuel conversion) are improved. In general, the gas yield and the H₂/CO ratio showed a constant value around 0.5, or even a very slight decrease, in the case of the biomass fuels tested at 1050 °C. On the contrary, the coal–coke blend showed both higher and increasing H₂/CO ratios when increasing the space residence time, which might be due to the combined effect of the catalytic enhancement of the water–gas shift reaction by some ash elements (Fe, Ni, Zn, and Al) and longer space times. This indicates that if biomass is used to produce syngas for biofuel production, the latter would require an upgrading stage in order to adjust the H₂/CO ratio.

- The combined effect of higher reaction temperature and higher space residence time has a positive effect on the gasification process, leading to an upgrade of the gas composition and higher gasification efficiencies. However, the increase in the H₂/CO ratio when the space time increases has been observed just for a temperature above ~1000 °C, this ratio decreasing with t_r for lower temperature. On the other hand, the fuel particle size has a negligible effect on the H₂/CO ratio.
- All the biomass fuels tested showed a better behaviour as compared to coal–coke, since they led to higher quality producer gas and higher gasification efficiencies, due to their higher reactivity.

Acknowledgements

The Ministry of Education and Science of the Government of Castilla-La Mancha is gratefully acknowledged for their financial support through the GACOMBIO Research Project (reference PCI08-0063). The authors are also grateful to the companies ELCOGAS, ALVINEA, CESEFOR, ENEMANSA, and FACTOR VERDE for supplying the fuels tested. G. Aranda is indebted to the Spanish Ministry of Science and Innovation for a FPU Scholarship (ref. AP2007-02747).

References

- [1] International Energy Agency, World Energy Outlook, 2007 <http://www.iea.org/textbase/nppdf/free/2007/weo_2007.pdf>, last accessed July 2009.
- [2] Commission of the European Communities, Green paper – A European Strategy for Sustainable, Competitive and Secure Energy, COM, 2006 105 final.
- [3] World Energy Council, Europe's Vulnerability to Energy Crises, 2008 <<http://www.worldenergy.org/documents/finalvulnerabilityofeurope2008.pdf>>, last accessed July 2009.
- [4] Commission of the European Communities, An Energy Policy for Europe, COM, 2007 1 final.
- [5] IDAE, Ministerio de Industria, Comercio y Turismo, Gobierno de España, Energía de la biomasa, Manuales de Energías Renovables, 2007 in Spanish, <http://www.idae.es/index.php/mod.documentos/mem.descarga?file=/documentos_10374_Energia_de_la_biomasa_07_b954457c.pdf>, last accessed July 2009.
- [6] M. Lapuerta, J.J. Hernández, J. Rodríguez, Kinetics of devolatilisation of forestry wastes from thermogravimetric analysis, Biomass and Bioenergy 27 (2004) 385–391.
- [7] A. Haryanto, S.D. Fernando, L.O. Pordesimo, S. Adhikari, Upgrading of syngas derived from biomass gasification: a thermodynamic analysis, Biomass and Bioenergy 33 (2009) 882–889.
- [8] Commission of the European Communities, Biomass Action Plan, COM, 2005 628 final, <http://europa.eu/legislation_summaries/energy/renewable_energy/l27014_en.htm>, accessed July 2009.
- [9] Ministerio de Industria, Turismo y Comercio, Gobierno de España, Plan de Energías Renovables 2005–2010, 2005 in Spanish, <[http://www.idae.es/index.php/mod.documentos/mem.descarga?file=/documentos_PER_2005-2010_8_de_gosto_2005_Completo\(modificacionpag_63\)_Copia_2_301254a0.pdf](http://www.idae.es/index.php/mod.documentos/mem.descarga?file=/documentos_PER_2005-2010_8_de_gosto_2005_Completo(modificacionpag_63)_Copia_2_301254a0.pdf)>, last accessed July 2009.
- [10] IDAE, Ministerio de Industria, Turismo y Comercio, Gobierno de España, Memoria anual, 2007 in Spanish, <http://www.idae.es/index.php/mod.documentos/mem.descarga?file=/documentos_10992_Memoria_anual_2007_08_c611f2f7.pdf>, last accessed July 2009.
- [11] A.V. Bridgwater, The technical and economic feasibility of biomass gasification for power generation, Fuel 74 (1995) 631–653.
- [12] P.N. Sheth, B.V. Babu, Experimental studies on producer gas generation from wood waste in a downdraft biomass gasifier, Bioresource Technology 100 (2009) 3127–3133.
- [13] C. Serrano, J.J. Hernández, C. Mandilas, C.G.W. Sheppard, R. Woolley, Laminar burning behaviour of biomass gasification-derived producer gas, International Journal of Hydrogen Energy 33 (2008) 851–862.
- [14] M. Lapuerta, J.J. Hernández, J. Rodríguez, Comparison between the kinetics of devolatilisation of forestry and agricultural wastes from the middle-south regions of Spain, Biomass and Bioenergy 31 (2007) 13–19.
- [15] Gasification Technologies Council, Gasification. Redefining Clean Energy, 2008 <http://www.gasification.org/Docs/Final_whitepaper.pdf>, last accessed July 2009.
- [16] J.J. Hernández, M. Lapuerta, C. Serrano, Estimation of the laminar flame speed of producer gas from biomass gasification, Energy and Fuels 19 (2005) 2172–2178.
- [17] J.J. Hernández, C. Serrano, J. Pérez, Prediction of the autoignition delay time of producer gas from biomass gasification, Energy and Fuels 20 (2006) 532–539.
- [18] IDAE, Ministerio de Industria, Turismo y Comercio, Gobierno de España, Biomasa: Gasificación, 2007 in Spanish, <<http://www.idae.es/index.php/mod.publicaciones/mem.detalle/reلمenu.73/id.35>>, last accessed July 2009.
- [19] M. Lapuerta, J.J. Hernández, A. Pazo, J. López, Gasification and co-gasification of biomass wastes: effect of the biomass origin and the gasifier operating conditions, Fuel Processing Technology 89 (2008) 828–837.
- [20] A. van der Drift, H. Boerrigter, B. Coda, M.K. Cieplik, K. Hemmes, Entrained flow gasification of biomass. Ash behaviour, feeding issues, and system analyses, 2004 ECN-C-04-039.
- [21] L. Wei, S. Xu, L. Zhang, C. Liu, H. Zhu, S. Liu, Steam gasification of biomass for hydrogen-rich gas in a free-fall reactor, International Journal of Hydrogen Energy 32 (2007) 24–31.
- [22] L. Wei, S. Xu, L. Zhang, H. Zhang, C. Liu, H. Zhu, S. Liu, Characteristics of fast pyrolysis of biomass in a free fall reactor, Fuel Processing Technology 87 (2006) 863–871.
- [23] K. Maniatis, Progress in biomass gasification: an overview, http://ec.europa.eu/energy/renewables/studies/doc/bioenergy/0000_km_tyrol_tony.pdf, last accessed July 2009.
- [24] HM Associates Inc., Princeton Energy Resources International, LLC, TFB Consulting, Assessment of the commercial potential for small gasification combined cycle and fuel cell systems, Phase II Final Draft Report for US DOE, 2003.
- [25] T.B. Reed, A. Das, Handbook of Biomass Downdraft Gasifier Engine Systems, Solar Energy Research Institute, 1988 SERI/SP-271-3022.
- [26] P.M. Lv, Z.H. Xiong, J. Chang, C.Z. Wu, Y. Chen, J.X. Zhu, An experimental study on biomass air–steam gasification in a fluidized bed, Bioresource Technology 95 (2004) 95–101.
- [27] F.V. Tinaut, A. Melgar, J.F. Pérez, A. Horrillo, Effect of biomass particle size and air superficial velocity on the gasification process in a downdraft fixed bed gasifier. An experimental and modelling study, Fuel Processing Technology 89 (2008) 1076–1089.
- [28] G. Chen, J. Andries, Z. Luo, H. Spliethoff, Biomass pyrolysis/gasification for product gas production: the overall investigation of parametric effects, Energy Conversion and Management 44 (2003) 1875–1884.
- [29] S. Rapagnà, A. Latif, Steam gasification of almond shells in a fluidised bed reactor: the influence of temperature and particle size on product yield and distribution, Biomass and Bioenergy 12 (1997) 281–288.
- [30] J.M. Encinar, J.F. González, J. González, Steam gasification of *Cynara cardunculus* L.: influence of variables, Fuel Processing Technology 75 (2002) 27–43.
- [31] J.-S. Chen, W.W. Gunkel, Modelling and simulation of co-current moving bed gasification reactors – part 1. A non-isothermal particle mode, Biomass 14 (1987) 51–72.
- [32] F. Mermoud, F. Golfier, S. Salvador, L. Van de Steene, J.L. Dirion, Experimental and numerical study of steam gasification of a single charcoal particle, Combustion and Flame 145 (2006) 59–79.
- [33] B.M. Jenkins, L.L. Baxter, T.R. Miles Jr., T.R. Miles, Combustion properties of biomass, Fuel Processing Technology 54 (1998) 17–46.
- [34] Y. Wang, C.M. Kinoshita, Kinetic model of biomass gasification, Solar Energy 51 (1993) 19–25.
- [35] G. Xu, T. Suda, Y. Matsuzawa, H. Tani, T. Fujimori, T. Murakami, Some process fundamentals of biomass gasification in dual fluidized bed, Fuel 86 (2007) 244–255.
- [36] O. Font, X. Querol, F. Plana, P. Coca, S. Burgos, F. García-Peña, Condensing species from flue gas in Puertollano gasification power plant, Spain, Fuel 85 (2006) 2229–2242.
- [37] O. Font, N. Moreno, S. Díez, X. Querol, A. López-Soler, P. Coca, F. García-Peña, Differential behaviour of combustion and gasification fly ash from Puertollano power plants (Spain) for the synthesis of zeolites and silica extraction, Journal of Hazardous Materials 166 (2009) 94–102.
- [38] CEN-TS 15104, Solid biofuels – determination of total content of carbon, hydrogen and nitrogen – instrumental methods, 2005.
- [39] CEN-TS 15289, Solid biofuels – determination of total content of sulphur and chlorine, 2006.
- [40] UNE 164001 EX, Biocombustibles sólidos. Método para la determinación del poder calorífico, 2005 in Spanish.
- [41] O. Senneca, Kinetics of pyrolysis, combustion and gasification of three biomass fuels, Fuel Processing Technology 88 (2007) 87–97.
- [42] E. Kastanaki, D. Vamvuka, A comparative and kinetic study on the combustion of coal–biomass char blends, Fuel 85 (2006) 1186–1193.
- [43] C. Di Blasi, F. Buonanno, C. Branca, Reactivities of some biomass chars in air, Carbon 37 (1999) 1227–1238.
- [44] F. Suárez-García, A. Martínez-Alonso, M. Fernández Llorente, J.M.D. Tascón, Inorganic matter characterization in vegetable biomass feedstocks, Fuel 81 (2002) 1161–1169.
- [45] J.M. Jones, L.I. Darvell, T.G. Bridgeman, M. Pourkashanian, A. Williams, An investigation of the thermal and catalytic behaviour of potassium in biomass combustion, Proceedings of the Combustion Institute 31 (2007) 1955–1963.
- [46] B.V. Babu, A.S. Chaurasia, Modeling for pyrolysis of solid particle: kinetics and heat transfer effects, Energy Conversion and Management 44 (2003) 2251–2275.
- [47] D.P. Ye, J.B. Agnew, D.K. Zhang, Gasification of a South Australian low-rank coal with carbon dioxide and steam: kinetics and reactivity studies, Fuel 77 (1998) 1209–1219.

- [48] D. Sutton, B. Kelleher, J.R.H. Ross, Review of literature on catalysts for biomass gasification, *Fuel Processing Technology* 73 (2001) 155–173.
- [49] UNE-CEN/TS 14775 EX, Biocombustibles sólidos. Método para la determinación del contenido en cenizas, 2004 in Spanish.
- [50] Johnson Matthey Catalysts, Katalco. High temperature shift catalysts, 2005 Commercial brochure. <<http://www.jmcatalysts.com/ptd/pdfs-uploaded/HTS%20Product%20Brochure.pdf>>, last accessed July 2009.
- [51] Johnson Matthey Catalysts, Katalco. Low temperature shift catalysts, 2005 Commercial brochure. <<http://www.jmcatalysts.com/ptd/pdfs-uploaded/LTS%20KATALCO.pdf>>, last accessed July 2009.
- [52] P. Lv, Z. Yuan, C. Wu, L. Ma, Y. Chen, N. Tsubaki, Bio-syngas production from biomass catalytic gasification, *Energy Conversion and Management* 48 (2007) 1132–1139.
- [53] S.C. Gebhard, D. Wang, R.P. Overend, M.A. Paisley, Catalytic conditioning of synthesis gas produced by biomass gasification, *Biomass and Bioenergy* 7 (1994) 307–313.